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THE QUESTION OF THE MECHANISM OF THE CRYSTALLIZING ACTION OF SO--ETC(U)
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THE QUESTION OF THE MECHANISM OF THE CRYSTALLIZING ACTION OF SOLIDS ON SUPERCOOLED WATER AEROSOLS

by

M. V. Tovbin, G. G. Buderaskaya

FOREIGN TECHNOLOGY DIVISION

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GREEK ALPHABET

Alpha	Α α α		Nu	Ν ν	
Beta	Β β		Xi	Ξ ξ	
Gamma	Γ γ		Omicron	Ο ο	
Delta	Δ δ		Pi	Π π	
Epsilon	Ε ε ϵ		Rho	Ρ ρ ϱ	
Zeta	Ζ ζ		Sigma	Σ σ ς	
Eta	Η η		Tau	Τ τ	
Theta	Θ θ ϑ		Upsilon	Υ υ	
Iota	Ι ι		Phi	Φ φ ϕ	
Kappa	Κ ϰ κ ϰ		Chi	Χ χ	
Lambda	Λ λ		Psi	Ψ ψ	
Mu	Μ μ		Omega	Ω ω	

RUSSIAN AND ENGLISH TRIGONOMETRIC FUNCTIONS

Russian	English
sin	sin
cos	cos
tg	tan
ctg	cot
sec	sec
cosec	csc
sh	sinh
ch	cosh
th	tanh
cth	coth
sch	sech
csch	csch
arc sin	$\sin^{-1}$
arc cos	$\cos^{-1}$
arc tg	$\tan^{-1}$
arc ctg	$\cot^{-1}$
arc sec	$\sec^{-1}$
arc cosec	$\csc^{-1}$
arc sh	$\sinh^{-1}$
arc ch	$\cosh^{-1}$
arc th	$\tanh^{-1}$
arc cth	$\coth^{-1}$
arc sch	sech^{-1}
arc csch	csch^{-1}

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rot	curl
lg	log

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THE QUESTION OF THE MECHANISM OF THE CRYSTALLIZING ACTION OF SOLIDS ON SUPERCOOLED WATER AEROSOLS.

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Kiev State University im. T. G. Shevchenko.

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Many researchers over a number of years conduct work on research on the mechanism of the crystallizing action/effect of different substances on the supercooled water aerosols. With the investigation of new reagents for an active effect on supercooled clouds and fog decisive importance has the laboratory evaluation of the effectiveness of synthesized specimen/samples, which precedes work consuming and bulky field tests. One should consider also that by the contemporary state of problem the investigation of new active preparations conducts empirically. As is known, until this time sole criterion for the preliminary selection of substances is the resemblance of crystal lattices of ice and crystallizing substance, which, as it shows experiment, not always is justified, but main, does not make it possible to obtain even the approximately

quantitative characteristic of the activity of substance. Thus, the selection of the crystallizing substances conducts by the mass tests.

The ice-forming activity of substance is characterized by the threshold temperature and the outcrop of the crystallization nuclei at the given temperature of the supercooled fog from the calculation on 1 g of reagent. Experimentally the threshold temperature determined from the appearance of single crystalline particles of ice in the supercooled fog. The outcrop of the crystallization nuclei significantly depends on the method of the dispersion of reagent and conditions of experiment.

Is suggested many procedures for determining the threshold temperature of ice formation. In Soviet investigations they put to use most frequently the cooling chambers of large volume [6]. In earlier work was applied cooling box [11, 13], sometimes Wilson cloud chamber [4]. In recent years is proposed the chamber of the original construction with strict thermostating the measure of the original construction with strict thermostating in volume [3], and also the procedure of cooling shaft/mine with lapse rate [2]. The most widely used methods of applying the chambers during ass testing specimen/samples are inconvenient due to unwieldiness and labor expense of experiments. Inconvenience is also that the tested substance must be introduced into the chamber in highly dispersed

state. Under laboratory conditions are always feasible the operations of the conversion of specimen/sample into aerosol by sublimation, fragmentation with explosion, etc. Finally, the application/use of the chambers is impeded by the need thoroughly to clean them from the reagents, which were being applied in the preceding/previous experiments. However, in spite of the deficiency/lacks indicated, it should be noted that these methods give thus far the only possibility of the estimation of the outcrop of the crystallization nuclei under conditions, which approximately imitate real cloud.

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In the present investigation was the expressed goal to develop the convenient method of research on the crystallizing action/effect of reagents and under identical conditions to compare the crystallizing action/effect of different substances. As the quantitative characteristic of the intensity of this action/effect can serve the caused by reagent increase in the temperature of the crystallization of the supercooled aerosol $\Delta T = T - T_0$ (where T and T_0 - crystallization temperature in the presence of reagent and without it). The procedure of the determination of value ΔT must satisfy some requirements. First, it must provide the possibility of obtaining the well reproducible results. In the second place, it is necessary that the conditions of experimentation as far as possible

be close to the themes, which are created in supercooled clouds. Taking into account that which was presented were carried out the investigations of the crystallizing action/effect of reagents on model systems. As the model of water aerosol in the developed method was utilized the emulsion of water in slightly polar viscosous liquid - liquid petrolatum.

In the literature there is information about the observation of the ice formation by optical method in reverse emulsions water/oil) [7]. For investigations the authors obtained the emulsion of water in liquid petrolatum, stabilized by the emulsifier T-2 (esterification product of maximum fatty acids and polyglycerol). For conducting our experiments we considered impossible the application/use of an emulsifier, since the addition of the latter could distort the picture of the determination of the temperature of the crystallization of water in the emulsion with the additions of reagent.

Experimentally by us studied the effect of a series of substances on the temperature of the crystallization of the supercooled emulsion of the water in oil, not containing stabilizers. For the purpose of an increase in the kinetic emulsion stability to liquid petrolatum they added certain quantity of carbon tetrachloride so that the generating mixture would have the same density as water.

This mixture, named oil, was utilized by us as the dispersive medium during the production of emulsion. The emulsion of water in oil was obtained by two different methods. In the first the emulsion was obtained by the dispersion of water in instrument for grinding biological preparations. For this, into the container of crusher poured 50 ml of oil and 1 ml of water. Mixture they mixed during 5 minutes at the velocity of the rotation of agitator 6000 r/min. As a result was obtained the uniform emulsion of water in oil, which, however, was understable and for several hours it was stratified. This, apparently, is explained themes that during the preparation of emulsion by means of energetic mixing into it unavoidably fall the air bubbles, which, floating, they carry off with themselves the droplets of water, causing their coalescence. In spite of this deficiency/lack, the obtained thus emulsion of water could be utilized for investigation, since the duration of conducting experiments was comparatively small (about 30 minutes).

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In the second method, based on the condensation reheated pair in cooled oil, were obtained the emulsions, kinetic more stable. For this into cooled to 0°C oil from fine/thin capillary during 15 seconds at constant velocity they passed the stream reheated water pair. In this case was obtained very stable in time emulsion with the

constant condensation of the dispersed phase (about 2o/o by volume), which was utilized for investigation.

It is interesting to note that data, obtained during the study of the temperature of the crystallization of the emulsions, prepared by the described two methods, will agree sufficiently well between themselves. For the characteristic of the applied emulsion, prepared by condensation method, was studied the particle distribution of the water in it according to size/dimensions. Figure 1 gives the spectrum of the drops of water in the emulsion. Along the axis of ordinates is deposit/postponed the number of particles (o/o) whose radius is less than the datum, along the axis of abscissas - the radius of particles r μm. As can be seen from the figure, the studied emulsions are polydisperse, with distinct maximum in distribution curve. In this case a greatest quantity of drops of water in the emulsio they have a radius 6 μm. These data show that this emulsion can serve as the sufficiently good model of natural water aerosols.

The temperature of the crystallization of emulsions was studied by thermograph method at the installation, schematically depicted on Fig. 2.

Into test tube A they poured 10 ml of emulsion and was placed into it copper-constantan thermocouple B. Test tube A on plug was

inserted into cooling chamber C whose walls were thermod-insulate by asbestos, through the chamber at the constant velocity, recorded by rheometer, they blow, cooled by liquid nitrogen, and through each of 30 seconds recorded readings of thermocouple B on a galvanometer of the type M-198/3. As an example Fig. 3 gives the typical thermogram, obtained in one of the experiments.

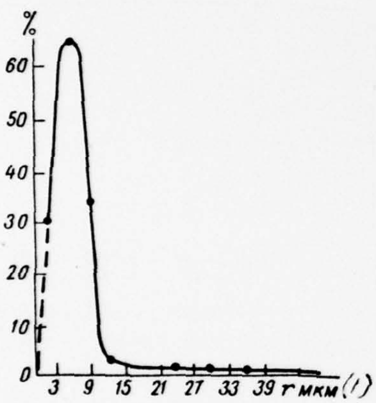

Fig. 1.

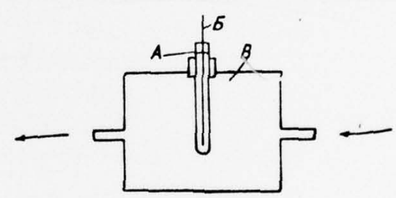

Fig. 2.

Fig. 1. Curve of the particle distribution of the water in the emulsion according to size/dimensions.

Key: (1). μm.

Fig. 2. Installation diagram.

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As can be seen from the figure, on thermogram is distinctly observable a series of the exothermic peaks (temperature, which corresponds to each of these peaks, it was determined from gauging curve/graph), caused by the freezing of the individual fractions of the drops of water, which were being accompanied heat liberation. The presence on the thermogram of a series of exothermic peaks is presented by completely natural, since the emulsions being investigated were polydisperse, and the temperature of th crystallization of the supercooled drops of water, as is known, it depends on their size/dimension. Furthermore, it is necessary to keep in mind that the process of the crystallization of supercooled liquid is fluctuational, and therefore even in the case of monodisperse systems on thermogram it was to be expected the presence of a series of peaks in the determined temperature range.

For the calculation of the most probable temperature of the crystallization of polydisperse emulsion, obviously, it is necessary to use statistical method. For this purpose they placed not less than 10 parallel experiments (so that on the obtained thermograms it would be about 100 exothermic peaks) and their results were treated statistically [4].

Figure 4 gives an example of the hystogram, constructed according to the empirical data of the freezing points of the drops of water in the emulsion, and the evened distribution curve according to the normal law of Gauss.

In this case probability that the empirical law corresponds to normal distribution, is 99.7o/o. With reliability 0.95 most probable the temperature of the crystallization of drops $\overline{T}$ = 17.3 ± 0.5°C.

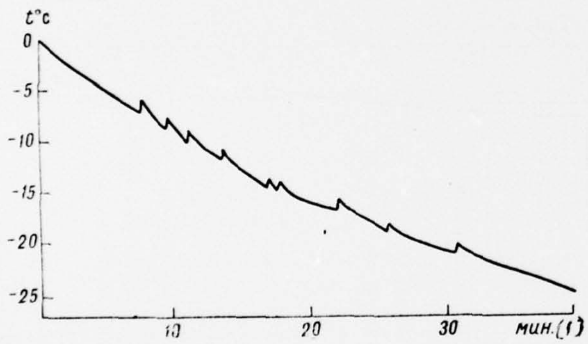

Fig. 3. Thermogram of the process of the crystallization of the supercooled emulsion of water.

Key: (1). min.

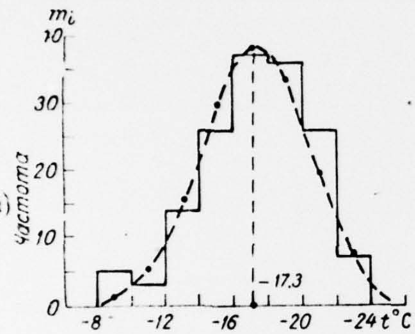

Fig. 4. Hystogram of the process of the crystallization of the supercooled emulsion of water.

Key: (1). Frequency.

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Then was initiated research on the effect of a series of substances on the temperature of the crystallization of water in the emulsion. In this case as the subjects of investigation were selected the reagents, possessing the considerable crystallizing action/effect

(AgJ, PbJ_2, CuS, phloroglucinol), and also for a comparison some substances, the not calling crystallizations of the supercooled aerosols ($AgCl$, CdS, PbS, $CaCO_3$).

As can be seen from Table 1, the results of the experiments, carried out with the emulsions of water, obtained by diff rent methods, they will agree sufficiently well between themselves. In accordance with literature data, the greatest crystallizing action/effect possess AgJ, PbJ_2, CuS and phloroglucinol. Such substances as $AgCl$, CdS, PbS, $CaCO_3$, either in practice do not affect the temperature of the crystallization of water, or even several it they lower ($CaCO_3$).

The given data also show that the proposed method can be used to evaluate and comparison of the crystallizing action/effect of different substances on the supercooled water aerosols.

By the very interesting to us is presented the established/installed experimentally fact of a sharp reduction in the emulsion stability of water in oil in the presence of a series of crystallizing reagents. This fact turned out to be that which was somewhat not expected. As is known, highly dispersed powders can be the stabilizers of emulsions. In this case hydrophobic powders (for example, carbon black) will stabilize reverse emulsions (of type"

water-in-oil"). As is known, the case of a reduction in the emulsion stability in the presence of highly dispersed powders, until now, are not described. The stabilization of emulsions by powders, and also the reverse process of a reduction in the emulsion stability, undoubtedly they present the surface phenomena, bonded with the accumulation of reagents on the boundary of two nonmiscible liquids.

Table 1.

(1) Вещество	(2) Эмульсия, полученная диспергационным методом		(3) Эмульсия, полученная конденсационным методом	
	T	ΔT	T	ΔT
(4) Вода	−17,3	—	−14	—
(5) AgJ	−8,5	8,8	−5,8	8,2
Флороглюцин	—	—	−6,8	7,2
PbJ_2	−9,6	7,7	−6,9	7,1
HgJ	—	—	−8,9	5,1
CuS	—	—	−9,3	4,7
CdS	—	—	−12,5	1,5
AgCl	—	—	−12,7	1,3
PbS	—	—	−14,4	−0,4
$CaCO_3$	−20	−2,7	−15,7	−1,7

Key: (1). Substance. (2). Emulsion, obtained by dispersion method. (3). Emulsion, obtained by condensation method. (4). Water. (5). Phloroglucinol.

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In connection by these it is very interesting to compare the action/effect of reagents with the character of its adsorptive reaction with water. This reaction can be studied by different methods, for example, by the study of the adsorption izotherms of

water vapors on this reagent, the determination of heat of wetting of solid with water, etc.

On the character of the reaction of the area of the particle of solid with water it is possible to judge also by their effect on the stability of aqueous emulsions. Therefore was compared crystallizing action/effect of reagents with their effect on the emulsion stability of water in oil.

For research on this question was utilized Rehbinder's method [8]. Experimentation technique was reduced to the following. Bunsen beaker was filled by the distilled water at the level of approximately 5 cm., and to it it was poured the same layer of liquid petrolatum. Into the layer of liquid petrolatum from fine/thin capillary was squeezed out the small drop of water (size/dimension of the drops of water in all experiments remained the constant and was equal to 0.05 ml), which slowly was omitted to interface water - oil. Experimentally determined time, after which occurred the coalescence of drop with the surface of water (the "lifetime" of drop - τ s.). The lifetime of drop, as is known, depends on many accidental experimental conditions, and therefore it is conducted 100 parallel measurements, which are subjected to mathematical processing [1], for the determination of most probable value τ.

For research on the effect of solids on emulsion stability instead of the water layer into beaker was poured the colloidal solution of the corresponding reagent. The obtained experimentally most probable values of the lifetime of the drop of water in the presence of the investigated reagents are given in Table 2.

As can be seen from table, some of the investigated substances cause a sharp decrease in the emulsion stability of water in oil, whereupon the lifetime of the drop of water decreases 3-4 times. This fact, as noted above, it is not expected. The mechanism of this destabilizing action/effect of powders requires supplementary study. Attracts to itself attention that that decrease the emuls on stability of water precisely those substances, which possess the considerable crystallizing action/effect on the supercooled water aerosols. Between the crystallizing action/effect of reagents, characterized by the found by us value ΔT, and by its ability to lower the stability of aqueous emulsion (which it is quan itatively can be evaluated by the value τ_0/τ, where τ_0 and τ are a lifetime of the drop of pure water, also, with the addition of reagents), as can be seen from Fig. 5, is observed sufficiently sharp correlation.

Table 2. Effect of solids on the emulsion stability of water in liquid petrolatum.

(1) Вещество . . .	(2) Вода	AgJ	PbJ_2	HgJ	CuS	AgCl	CaS	PbS	$CaCO_3$
(3) τ сек.	131	27	45	57	75	99	99	128	176

Key: (1). Substance. (2). Water. (3). τ, s.

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This derivation will agree with the views of a series of the researchers [9, 12, 15]. It is of considerable interest, since surface can be modified by adsorptive methods, which open/discloses the new ways of an artificial change in the crystallizing action/effect of reagents.

For the check of the last/latter position we conducted several preliminary experiments. In the first of them was hydrophylized surface AgJ. This was reached by the adsorption of the traces of gelatin by the highly dispersed suspensions AgJ. It turned out that after this adsorptive surface modification the crystallizing action/effect sharply descends. The possibility of an increase in the crystallizing action/effect of substances was checked on the highly dispersed powders of quartz. The quartz, which has the crystal structure, close to the structure of ice, nevertheless as showed the

experiments, it does not affect the temperature of crystallization of the supercooled emulsion. This is caused, apparently, the es that quartz has hydrophilic surface. After the partial waterproofing of the surface of quartz by processing its dimethyldichlorosilane it became to exhibit the sufficiently high crystallizing activity.

Conclusions.

1. Is described the method of the determination of the crystallizing action/effect of reagents, based on research on their effect on the freezing point of the supercooled emulsion of water in oil.

2. Is assumed that the crystallizing action/effect of reagents on the supercooled water aerosols is caused by the specific character of adsorptive processes on interface water - reagent.

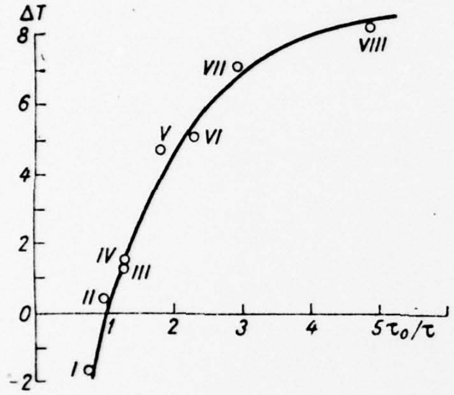

Fig. 5. Correlative dependence between the crystallizing action/effect of reagent and its ability to lower the emulsion stability of water in oil. I) $CaCO_3$, II) PbS, III) AgCl, IV) CdS, V) CuS, VI) HgS, VII) PbJ_2, VIII) AgJ.

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3. Is studied the effect of a series of solids in highly dispersed state on the emulsion stability of water in liquid

petrolatum. It is shown, that a series of solid reagents causes a sharp decrease in the emulsion stability.

4. It is establish/installed that between the ability of reagents to lower the emulsion stability of water and the r crystallizing action/effect on the supercooled water aerosols is a sharp correlative dependence.

5. The findings confirm assumption about the fact that the crystallizing action/effect of reagents is caused by the character of their surface reaction with water.

6. It is shown, that by the surface modification of reagent by adsorptive methods it is possible to affect its crystallizing action/effect.

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